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Majborn, Benny; Brodersen, K.; Damkjær, Anders; Højerup, C.F.

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Nuclear Safety Research Department Annual Progress Report 1994

Edited by B. Majborn, K. Brodersen, A. Damkjær and C.F. Højerup

Risø National Laboratory, Roskilde, Denmark
March 1995

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Abstract The report describes the work of the Nuclear Safety Research Department during 1994. The activities cover health physics, reactor physics, operation of the small reactor DR1, and radioactive waste management.

Lists of staff and publications are included together with a summary of the staff's participation in international committees.

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Contents

1 Introduction

The Nuclear Safety Research Department is engaged in research and development concerning radiation protection, reactor safety and radioactive waste. In 1994 the department was organized in three sections: The Health Physics Section, the Reactor Physics Section (with the reactor DR1) and the Waste Management Section.

In addition to its research and development activities, the department is involved in supporting activities related to the safe operation of the nuclear facilities at Risø. The activities include personnel dosimetry, maintenance and calibration of health physics instruments, emergency preparedness, reactor physics support to the reactor DR3, criticality evaluations, and participation in safety committee work. The Waste Management Section is responsible for the safe handling and storage of radioactive waste from Risø as well as from other Danish users of radioactive materials.

The work of the department involves a close collaboration with the Danish nuclear authorities: the Emergency Management Agency and the National Institute of Radiation Hygiene, and also with Danish and foreign universities and research institutes, especially the Technical University of Denmark and partners in research programmes supported by the EC (European Commission) and by NKS (the Nordic Nuclear Safety Research Programme).

As part of a change of organization at Risø it is expected that a new department of Nuclear Safety Research and Nuclear Facilities will be formed in March 1995. The new department will be composed of the Nuclear Safety Research Department, the Research Reactor DR3 and the Isotope Laboratory.

This report describes the work of the Nuclear Safety Research Department in 1994 with an emphasis on the results of the research and development activities. Lists of staff and publications are included together with a summary of the staff's participation in international committees.

2 Health Physics

The Health Physics Section works with research and development in dosimetry, instrumentation for radiation measurements, and radon physics. In addition the section is responsible for the personnel dosimetry and for the calibration and maintanence of the health physics instruments at Risø. The section also contributes to the Danish emergency planning and preparedness systems.

2.1 Dosimetry

2.1.1 Personnel Dosimetry

Risø's personnel dosimetry service covers the individual monitoring of the personnel at Risø and at the Niels Bohr Institute Tandem Accelerator. Only persons actually involved in radiation work are equipped with a personal dosemeter. In areas where the use of personal dosemeters is not required, the radiation levels are controlled through an extensive area-monitoring programme using thermoluminescense (TL) dosemeters.

The main statistics of the dosimetry service for 1994 are shown in Table 2.1 and in Figure 2.1.

Table 2.1 Statistics for monitoring of Risø personnel in 1994.

No. of persons monitored	873
No. of persons receiving external doses above 0.2 mSv (the registration level)	213
No. of persons receiving internal doses from intake of tritiated water, or intake of other radioactiviy	58 2
Total collective equivalent dose to the monitored personnel: External doses Internal doses Total	 262 mSv 15 mSv 277 mSv

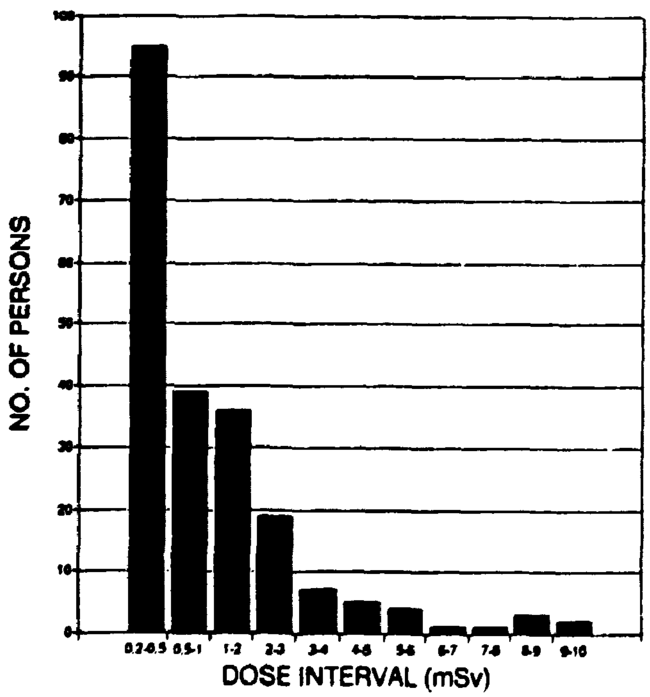

Figure 2.1. Distribution of whole-body doses (effective doses) for Risø personnel in 1994

A new PC-operated program for the administrative handling of the measured personal dose data has been developed and initially tested. The program is adapted for the new reporting procedures to the authorities and for new requirements for reporting internally at Risø. The program will be ready for use in 1995.

2.1.2 EU Technical Recommendations on Individual Monitoring

In collaboration with other European laboratories, the dosimetry group assisted the EU in preparing a document on technical recommendations for monitoring the exposure of individuals to external radiation. The final document has been published as Report EUR 14852 EN.

2.1.3 Dosimetry of Beta and Low-Energy Photon Radiations

Study of GM Counters for Dose Rate Measurements i Beta Radiation Fields

The aim of this project was to utilise the extremely high sensitivity of particle detectors, e.g. GM counters, for dose rate measurements in beta radiation fields where ionisation chambers have some shortcomings. As the conversion factor K_{Beta} for converting the measured count rate to dose rate is highly dependent on beta energy, some information on the energy of the beta radiation must also be provided by the measurement. In this work the possibility of using two different absorbers to provide useful information on the beta energy for obtaining a value of K_{Beta} was studied. The outcome of this study might be a dose-rate-meter construction consisting of two equal GM detectors provided with different absorbers.

The measurements were carried out with a LND-type GM tube (30 mm in diameter, and approximately 1 cm deep) with an end-window thickness equivalent to 1.60 mg/cm^2. By using a number of absorbers in front of the GM detector, absorption curves were obtained for beta radiation fields from the beta emitters ^{90}Sr/^{90}Y (E_{max} = 2.26 MeV), ^{204}Tl (E_{max} = 0.77 MeV), ^{147}Pm (E_{max} = 0.22 MeV) and ^{14}C (E_{max} = 0.155 MeV). Dose rate data obtained with an extrapolation chamber, which is the standard instrument for measurement of dose rates in beta radiation fields, were used to obtain values of K_{Beta} for the different beta radiation fields. In Figure 2.2 the K_{Beta} values for the measurement of skin dose rates, i.e. dose rates to tissue at a depth of 0.07 mm, are presented as a function of the ratio of the GM count rates measured with two absorbers, one without additional absorber to the end-window thickness of 1.6 mg/cm^2 and the other one with a total absorber thickness (window+filter) of 7 mg/cm^2. The figure clearly illustrates that for unknown beta radiation fields the ratio of GM measurements with two different absorbers can be used to obtain a value of K_{Beta}.

The method will be further studied for application for mixed beta and beta/photon fields as well as for different angles of incidence of the radiation.

Establishment of a Low-Energy Beta Calibration Facility

Due to the strong absorption in air of low-energy beta particles (1 cm air will attenuate the beta dose rate from a ^{63}Ni source with E_{max} = 67 keV by more than 90%) a significant degradation of the beta energy spectrum will be observed at short distances from the source. The requirement of an unmoderated beta spectrum

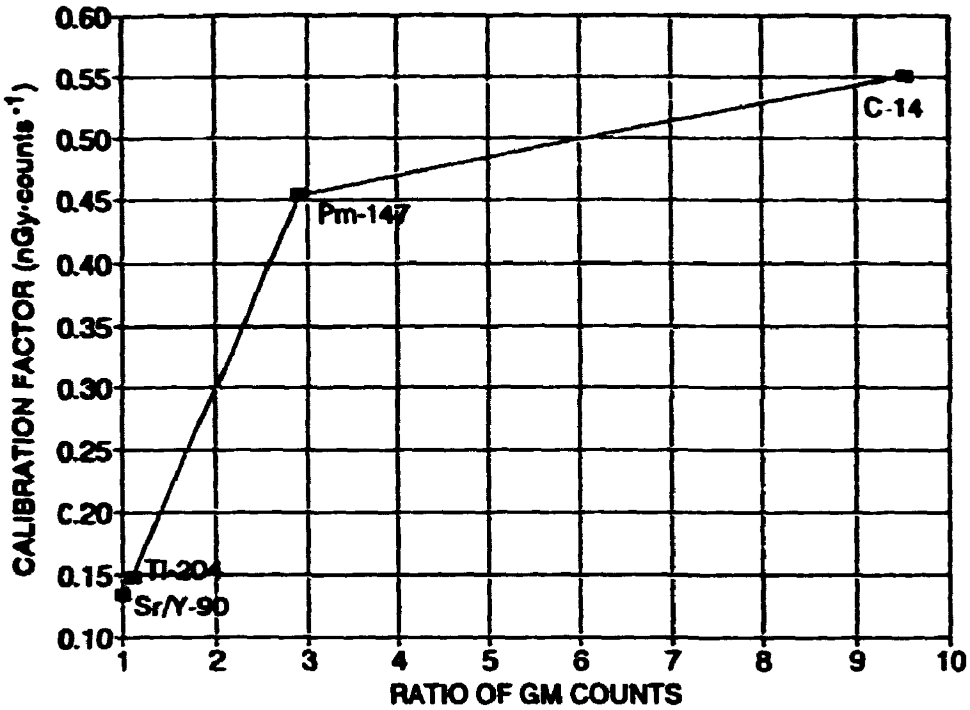

Figure 2.2. Beta dose calibration factor, $K_{tw,d}(0.07;0°)$, for the GM detector provided with 7 $mg.cm^{-2}$ total absorber, as a function of the ratio of measured GM counts for the two total absorbers, 1.6 and 7 $mg.cm^{-2}$.

for calibration purposes can therefore only be fulfilled if the irradiation is made very closely to the surface of the source, which gives rise to practical problems. An improved irradiation geometry can be obtained if the irradiations take place under reduced air-pressure conditions, allowing the irradiations to be made at larger distances with the beta energy spectrum being kept essentially unchanged.

For this purpose a container, of diameter 50 cm and length 35 cm, has been constructed, which can be evacuated, and in which the beta source can be established for irradiation under reduced air-pressure conditions. The development and construction of the chamber was made in collaboration with the Engineering and Computer Department. The chamber will be used for establishing a calibration facility for low-energy beta radiation, which will be useful for studying the response of thin detectors for irradiation with low-energy beta particles.

Establishment of a ^{55}Fe 6-keV Photon Calibration Facility

A 37 GBq ^{55}Fe source purchased from the Amersham Company has been arranged in an irradiation setup. The ^{55}Fe activity is plated on one face of a 13 mm diameter copper disk and covered by a 0.25 mm thick protective beryllium window. The photon energy spectrum of the source (shown in Figure 2.3) was obtained with a 80 mm^2 cooled silicon-lithium drifted planar detector with assistance from the Environmental Science and Technology Department.

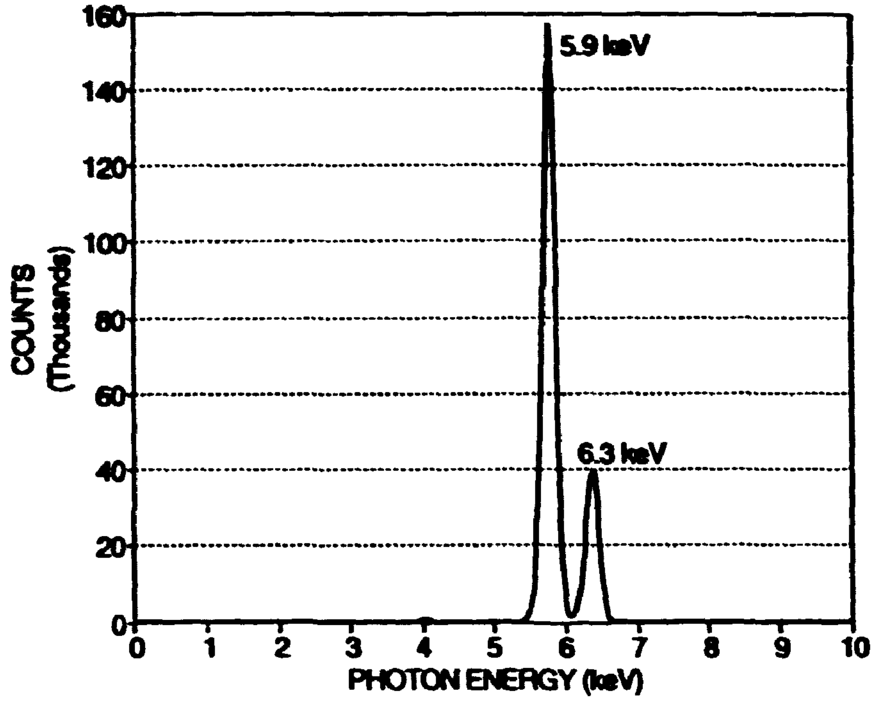

Figure 2.3. Measured photon energy spectrum from the established ^{55}Fe source.

The irradiation facility has been provided with an automated extrapolation chamber facility for characterising the radiation field from the source in terms of absorbed dose rate to tissue. The development and establishment of the extrapolation chamber facility was made in collaboration with the Engineering and Computer Department.

In 1995 the radiation field will be characterised in terms of exposure rate as well as dose rate, and conversion coefficients for converting from exposure to absorbed dose to tissue will established. The results will be compared with the results obtained at Fontenay-aux-Roses from similar measurements.

Study of Thin TL Detectors

The capability of thin TL detectors for measuring absorbed dose rate to tissue in beta radiation fields has been further studied. The energy and angular responses as well as sensitivity and detection threshold have been measured for different types of thin TL detectors. Generally an efficiency of 0.3-0.5 was observed for the measurement of beta doses from radiation fields from ^{14}C and ^{147}Pm, except for the carbon-loaded MgB_4O_7:Dy which showed an efficiency of up to 0.9. By using appropriate thicknesses of the filters in front of the detectors, dosemeters showing satisfactory accuracy for the measurement of $H_p(0.07)$ can be obtained.

Monte-Carlo (EGS4) Calculations and Validation in Beta Dosimetry

The Monte-Carlo computer code EGS4 was applied for modelling the beta and photon (Bremsstrahlung) energy spectra and dose rates produced by a 50 mm diameter ^{14}C source. The calculated data were experimentally validated by measurements with Si(Li)- and HPGe-detectors.

The work was carried out as part of a Ph.D. project at the Institute for National Measurement Standards of the National Research Council, Canada.

2.1.4 Control of Irradiated Spices

The dosimetry group assisted the FDB company with TL measurements of a number of selected samples of spices for irradiation control.

2.2 Development of Instruments and Methods

2.2.1 Retrospective Radiation Dose Reconstruction Using Optically Stimulated Luminescence on Natural Materials

Optically stimulated luminescence (OSL) techniques, especially aimed at using natural materials for retrospective reconstruction of accidental radiation doses in populated areas, were developed and studied at Risø as part of an EU research project. Quartz and feldspars separated from building materials, such as bricks and tiles, in addition to porcelain from toilet tanks, had their OSL signals measured using different light sources for stimulation to assess radiation doses received by the material. Radiation doses were also evaluated from OSL measured directly on unseparated samples, i.e. directly from the surface of brick and tile materials.

In an effort to apply optical stimulation for retrospective radiation dosimetry, a number of basic studies of OSL techniques were undertaken at Risø. The materials investigated include quartz and feldspars extracted from bricks, outdoor tiles, and porcelain from toilet tanks.

Apparatus and Techniques

The apparatus used for the experimental work were mainly OSL units developed as attachments to the automated Risø TL-reader and include a scanning monochromator for wavelength-resolved luminescence measurements. An instrument for continuous OSL scanning of sediment cores was also developed. The latter technique naturally lends itself to continuous scanning measurements of brick cross-sections, allowing depth dose profiles to be measured directly.

The basic OSL unit, containing light sources for both green light and infrared stimulation, enables measurements of OSL signals from both quartz and feldspar samples. Green light stimulated luminescence (GLSL) is achieved by illumination with a filtered light spectrum from a halogen lamp using exchangeable excitation and detection filter packs. The GLSL unit is designed to select a green light stimulation wavelength band using excitation filters extending to as low a wavelength as possible while still being sufficiently separated from the luminescence emission spectrum. The infrared stimulated luminescence (IRSL) is generated by an infrared diode array (peak emission of 875 Δ 80 nm) placed close to the sample. While GLSL can be used with both quartz and feldspars, IRSL seems only to work with feldspars.

Ideally, however, the spectral excitation and emission characteristics of quartz and feldspar materials prepared for dosimetric evaluation would be routinely scanned, since this would also allow the possibility of choosing the most suitable energy windows in which to carry out the measurements. A compact module was developed that allows for the monochromatic illumination of samples in the wavelength range 380 to 1020 nm, enabling the measurement of energy resolved OSL. The unit can be directly coupled to the existing automated Risø TL/OSL system. The unit can also be used for recording wavelength-resolved emission spectra, whether photo excited or thermally stimulated. A schematic diagram of the combined OSL attachment is shown in Figure 2.4.

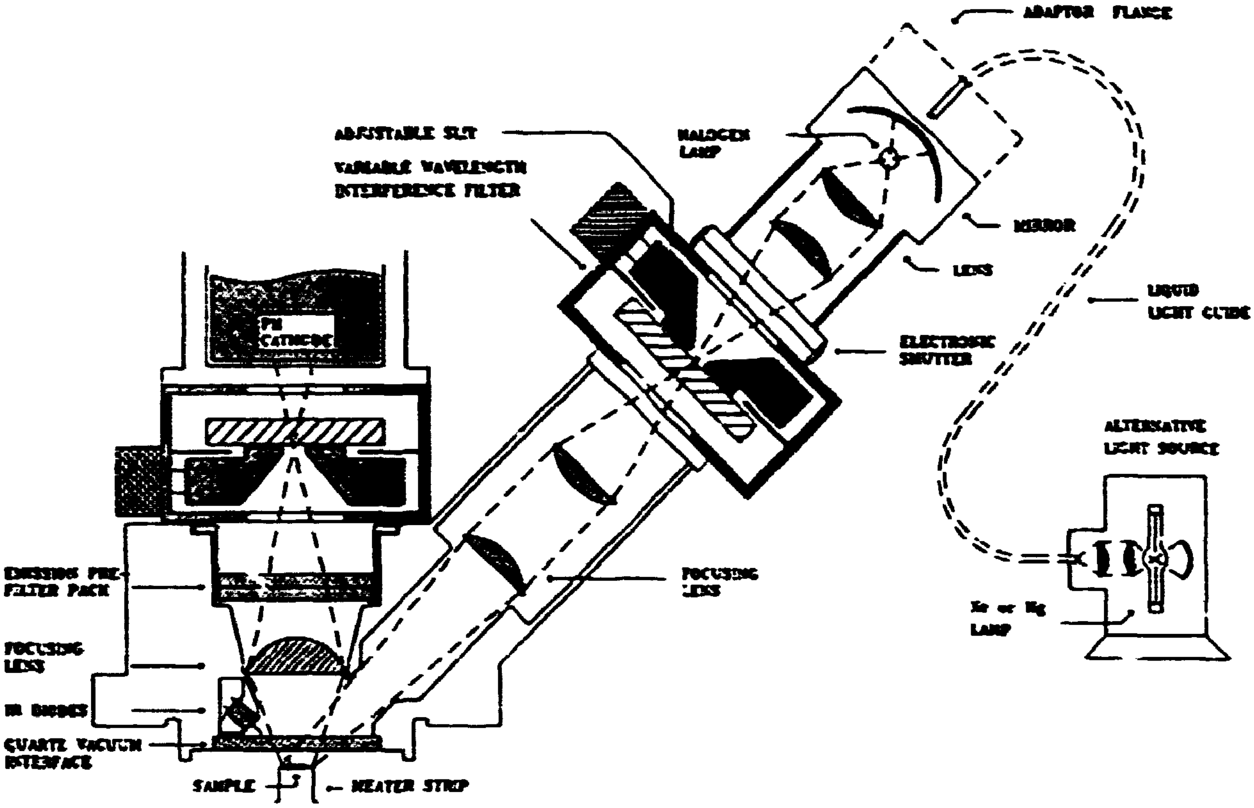

Figure 2.4. Schematic diagram of the OSL attachment showing the excitation lamp system with monochromators mounted both on the excitation side and the detection side.

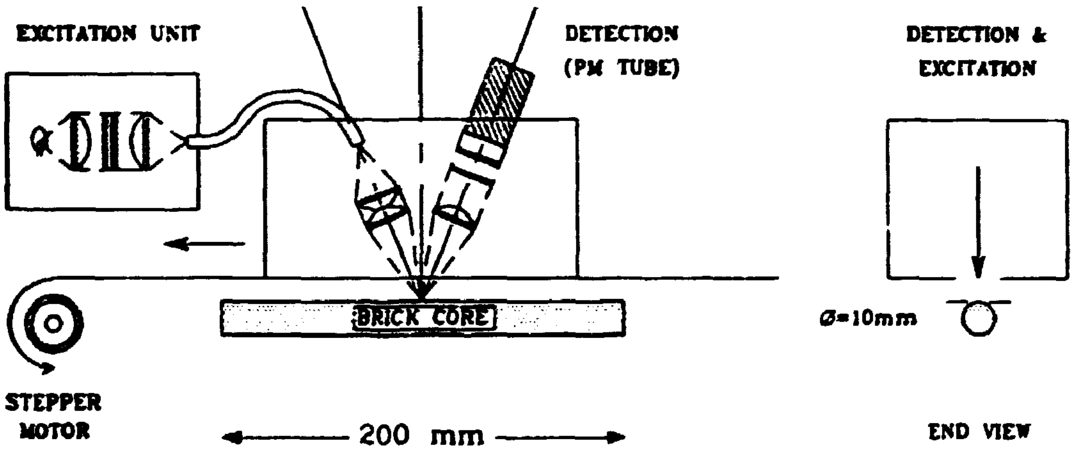

Figure 2.5. Schematic diagram of the automatic OSL core scanning system: the excitation beam is 1 mm wide.

The continuous OSL core-scanner system allows the optical sensors to be moved across either sediment or brick cores. A stepper-motor drive ensures constant scan rates and accuracy in positioning to better than 0.1 mm. The optical sensor system was developed at Risø and consists of a photo-excitation and detector module together with lamps for bleaching and regenerating OSL. Photo excitation is made using a filtered halogen lamp generating a green wavelength band (420-550 nm) and the brick core is scanned using an excitation slit beam of 10 mm x 1 mm which determines the resolution of the system. Photo detection is made through a 6 mm standard Hoya U-340 filter (peak emission at 340 nm). OSL dose normalization is made either by using short-wave UV light from a 20 W low-pressure HG lamp or exposing the brick cores to a Cs-137 gamma field and afterwards scanning the OSL sensitivity across the brick profile. A schematic diagram of the OSL scanner system is shown in Figure 2.5.

Results

The OSL sensitivity of quartz depends very much on the thermal history of the material with fired samples giving up to an order of magnitude larger signals than non-heated samples. An attempt to determine the lower detection levels using green light stimulated luminescence on fired quartz was made by obtaining dose versus GLSL response curves for a variety of quartz samples extracted from archaeological specimens such as bricks, burnt stones and clay. The GLSL response curve for a sensitive quartz extracted from burnt clay, obtained using the multiple-aliquot method, is shown in Figure 2.6 A. As can be seen, the lowest detectable dose for this material is well below 1 mGy.

A single-aliquot method was also tested on the same quartz sample as above where the dose response curve was obtained using the regeneration technique. Beta irradiations were carried out using the Sr-90 source of the Risø TL/OSL reader and GLSL measurements were carried out as described above. The results obtained show very little scatter because of the single-aliquot technique used, where no normalization is needed. Figure 2.6 B presents the dose versus GLSL response curve in the dose range 0.1 to 5 mGy compared with that of the TL response. The lower detection level using GLSL on this particular sample is seen to be well below 0.5 mGy.

The accumulated "natural" dose induced in quartz by radionuclides contained in modern brick materials and by the environmental gamma radiation was measured using GLSL on extracted quartz samples. This experiment was aimed at deter-

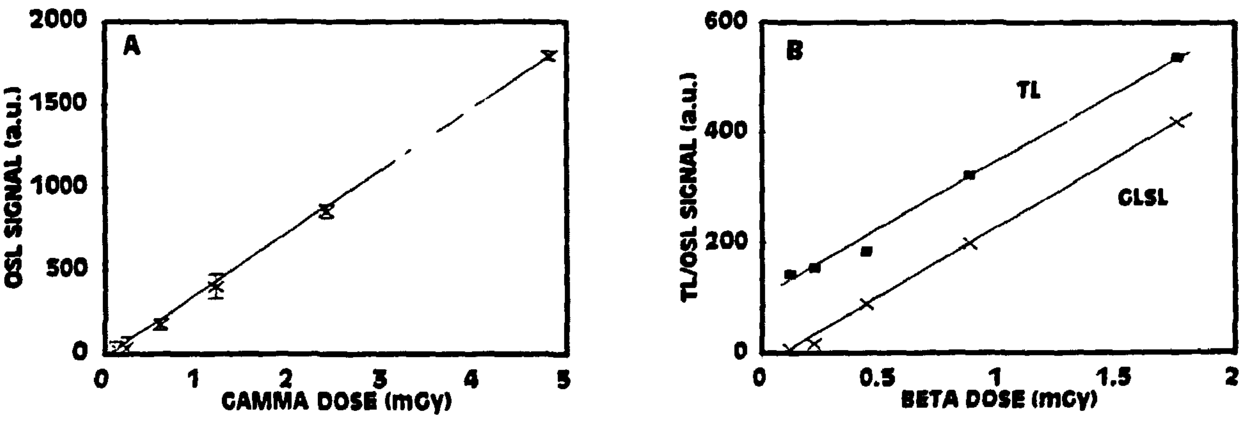

Figure 2.6. (A) OSL versus Co-60 gamma dose (multi-sample technique) for quartz extracted from a burnt stone. (B) TL and OSL versus beta dose (single-aliquot method) for the same quartz sample.

mining the lower detection limit for an additional dose received by a brick as a result of radioactive release from a nuclear accident taking into account the GLSL contribution from the natural background dose. The quartz grains were extracted from the material and the absorbed dose was determined by GLSL using the additive dose technique. The dose was estimated to be about 200 mGy which is in very good agrement with the expected value based on an annual dose rate of about 5 mGy/y from the environmental radiation and the natural radioactivity in the brick. For this particular brick the lower detection limit for an additional accidental dose would be of the order of 20 mGy (10% above the background).

Depth-dose profiles in bricks can be determined by measuring the OSL signals directly from the unseparated material across the brick. Modern bricks were annealed at 500°C to remove any previously acquired TL/OSL signal and then exposed to Co-60 and Cs-137 photon radiation fields, respectively. After irradiation, 8 mm diameter cores were drilled out of the brick and sliced into 1 mm thick circular discs using a diamond saw. Each disc, representing a particular depth in the brick, had their GLSL measured directly from the surface of the unseparated material, and as an example the OSL versus depth for an ancient brick irradiated with Co-60 radiation is shown in Figure 2.7 A. As can be seen, the half-value layer is about 75 mm, which corresponds reasonably well with the expected attenuation of Co-60 photons in brick material. The measured doses as function of depth into the brick material for ^{137}Cs and ^{60}Co irradiation geometries are shown in Figure 2.7 A and Figure 2.7 B, respectively.

The automatic core scanner system was used to measure the depth-dose profiles across the 200 mm length of a modern brick exposed in the laboratory to Co-60 and Cs-137 gamma radiation doses, respectively. 10 mm cores were drilled from the brick after irradiation and mounted directly under the light sensing head of the core scanner. The cores were scanned with a speed of 1 mm per sec. using an excitation light density of 20 mW/cm2. Normalization was made after bleaching the cores either by exposing the cores by UV light produced by the attached low-pressure Hg lamp or by exposing the cores perpendicularly to a Cs-137 gamma field. Figures 2.8 A and 2.8 B show the depth-dose profiles obtained from OSL scanning of cores from a brick that had been exposed from one direction in the

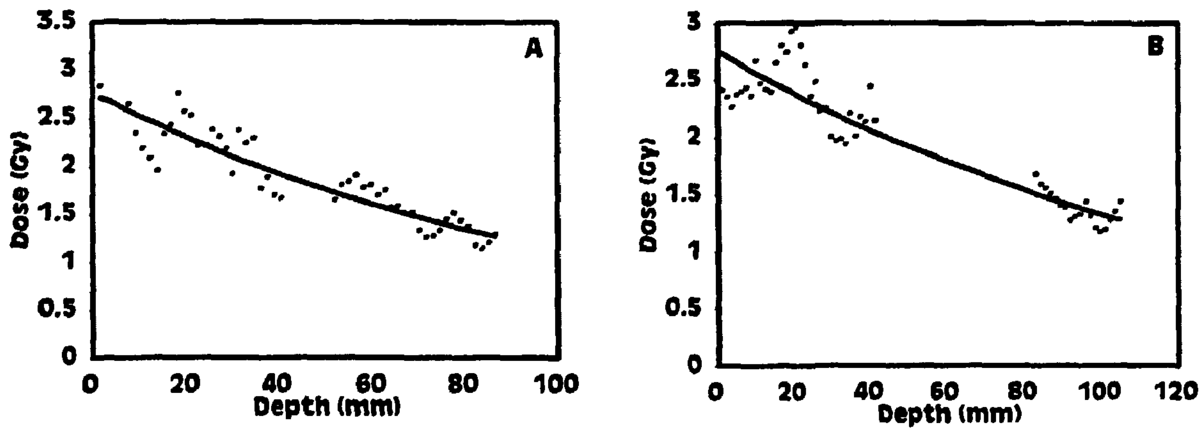

Figure 2.7. Dose versus depth into a brick irradiated with gamma radiation from one side. Measurements were made using green light OSL on 8 mm diameter x 1 mm slices cut from a core through the brick. (A) Profile for 5 Gy ^{137}Cs gamma radiation. (B) Profile for 5 Gy ^{60}Co gamma radiation.

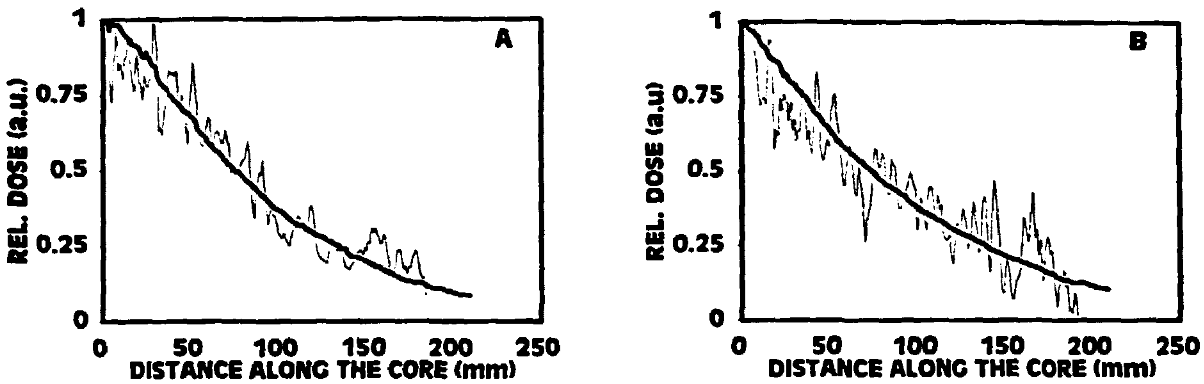

Figure 2.8. Relative depth-dose profile into a brick from (A) ^{137}Cs and (B) ^{60}Co gamma radiation from one side as calculated by the Monte Carlo code MCNP (bold lines). For comparison, the relative depth-dose profiles measured with the automatic OSL core scanner system are also shown.

laboratory to Cs-137 and Co-60 radiation, respectively (20 Gy). Monte Carlo calculated attenuation curves for the same irradiation geometries are shown as well, and they compare well with the experimentally obtained curves.

Finally, an assessment was made as to whether the OSL technique can be used as an alternative method to the established thermoluminescence technique for measuring accrued dose levels in porcelain: since no heating is required, problems of heat induced sensitivity changes during measurement are overcome. The OSL versus beta dose for a porcelain sample was found to increase linearly with dose in the range 0-16 Gy and shows a further sublinear increase of OSL signal up to at least 200 Gy. These preliminary measurements indicate that dose levels of about 0.1 Gy can be determined using OSL techniques on porcelain.

A Ph.D. project on retrospective dosimetry using OSL on natural materials was initiated in 1994 and this project has mainly been concentrated on investigations of OSL sensitivity changes in quartz as a result of annealing at high temperatures. Heated quartz materials gives order of magnitudes higher OSL signals than that of non-heated quartz. This is crucial when using e.g. extracted quartz from building materials (bricks) that were heated in the past. Also OSL sensitivity changes of quartz as a function of re-use (regeneration method) has been studied and some results have been presented at the international LUMDETR'94 conference in Tallinn, Estonia (September 1994).

2.3 Radon Research

Radon Test Structure

The radon test structure is an experimental facility for field studies of transport of soil gas and radon through soil and entry into houses. The structure provides a well-defined building shell similar to a small slab-on-grade house (2.6 x 2.6 m in floor area), where the entry process can be studied without the influence of house occupants or uncontrolled changes in ventilation.

During 1994, the experimental work has focused on the mapping of radon concentration fields below the structure when air is forced into the ground below the structure. These experiments simulates the mitigation technique known as subslab pressurization. Figure 2.9 shows two radon concentration fields obtained during the summer of 1994. The top plot is with an air flow of 18 litres per min being forced into the ground, and the bottom one is without any flow (diffusive mode). Each circle on the plots represents two to fc , probe locations in the ground from where soil gas has been sampled and analysed for radon. As can be seen, the soil in the vicinity of the structure is strongly depleted for radon when the flow is applied. The degree of radon depletion depends directly on the transport properties of the soil matrix and fractures. Radon concentration fields obtained under a range of experimental conditions are presently being compared with results of numerical modelling based on combined diffusive and advective transport.

The soil-gas entry rate into the test structure for a given depressurization has been measured during the fall and winter seasons of 1994. As can be seen in Figure 2.10, the soil-gas entry rate is strongly influenced by the depth of the water table. This indicates that the flow into the test structure is not merely confined to single fractures and cracks (e.g. along the interface of the concrete foundation) but involves the entire soil matrix down to the water table. A set of numerical model calculations in two dimensions based on the assumption that the

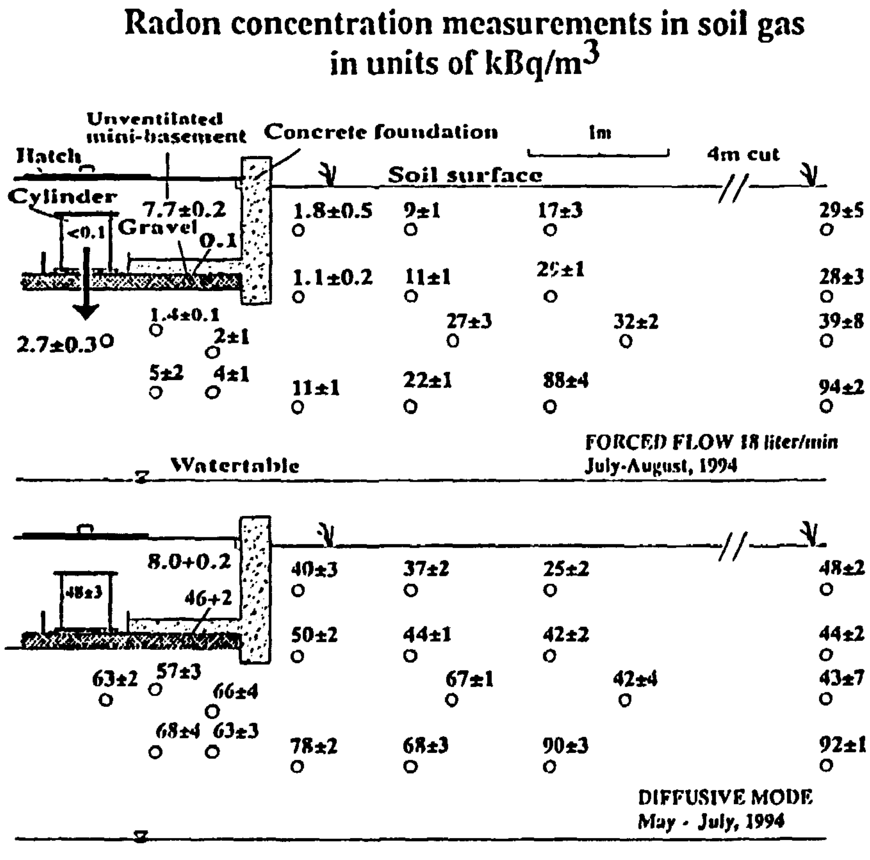

Figure 2.9. Radon concentration fields below the test structure. In the plot to the left, radon-free air is forced into the ground at a rate of 18 litres per minute. In the plot to the right, no forced flow is present, and only natural driving forces are in operation.

soil is homogeneous and has a constant content of moisture (i.e. independent of the depth of the water table) confirms that the relationship may be explained primarily as a geometrical effect due to the capping off of the bottom soil.

In 1994, the site characterization was completed. The Danish Geotechincal Institute (GI) drilled 6 boreholes of 15 cm in diameter, and undisturbed soil samples were taken every 15 cm from the surface of the soil to a depth of 3 m. 4 samples per borehole were analysed by GI for porosity, contents of moisture, and soil density. Part of the remaining samples are currently being analysed for radon emanation in the laboratory.

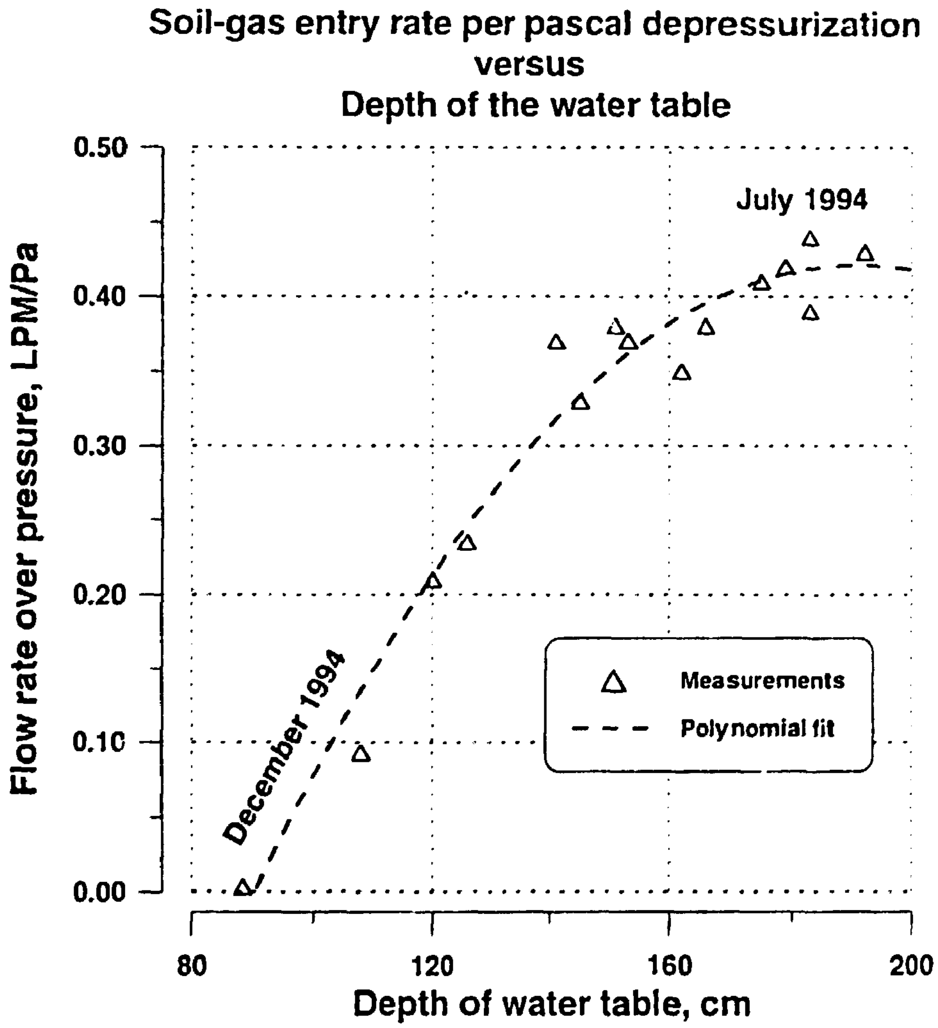

Figure 2.10. Soil-gas entry into the test structure.

Radon Transport in Fractured Media

In 1994, a Ph.D. project on transport of radon in inhomogeneous and/or fractured media was initiated. The work has focused on the design and planning of soil-column experiments using large intact samples of clayey till collected by the Geotechnical Institute. The plan is to map break-through curves of radon under a range of experimental conditions, and from that to deduce the importance of fractures and matrix transport properties.

Instrumentation

Two multicounters that each handles 12 scintillation cells were completed and put into operation. Special software that controls the multicounters and acts as a database and report writer were completed. The database presently contains more than 3000 single scintillation cell measurements. Ten new scintillation cells were fabricated in the workshop and are now used for low-level counting such as radon emanation measurements. Two sets of absolute radon standard sources are under preparation. These sources (or bubblers) are based on standard radium solutions from the National Institute of Standards and Technology (USA), and chambers of known volumes. The standard sources and a reference instrument based on a surface barrier detector will be completed primo 1995.

2.4 Emergency Preparedness

On May 17 the Technical Emergency Response Centre (TBT) at Risø together with the Emergency Management Agency (EMA) and a number of Swedish participants took part in the exercise YPSILON.

The exercise scenario started with an airplane crash in a storage building at the Barsebäck Nuclear Power Plant. All the measurement results reported during the exercise were calculated in advance by means of a PC-version of the Risø code PLUCON.

The sensitivity of the National Radiation Monitoring Network were greatly improved after the introduction of new measured standard spectra for radon daughters and other naturally occurring isotopes.

The contract with the Emergency Management Agency (EMA) concerning transfer of software for the National Radiation Monitoring Network from UNIX to WINDOWS NT is finished. The data are now continously collected on a NT-system as well as on the old UNIX-system. In order to test the system, three computers are now running WINDOWS NT connected through the Risø ethernet and through a 64 kbit/s line with the EMA.

2.5 Publications

2.5.1 Publications in International Journals, Proceedings and Reports

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Walmod-Larsen, O., Intervention principles and levels in the event of a nuclear accident. In: Beredskap mot atomulykker. Nordisk seminar beredskap mot atomulykker, Oslo (NO), 4-5 May 1994. (Nordisk Kjernesikkerhetsforskning, Sandvika, 1994) 7 p.

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Unpublished. Abstract available

3 Reactor Physics

Work in the Reactor Physics Section falls mainly in the three categories

1) Reactor Physics
2) Reactor Safety
3) Activities at the DR 1 reactor

3.1 Reactor Physics

Reactor physics is the discipline concerned with the nuclear processes in reactor cores and as such fundamental for the understanding of both operation and safety aspects of nuclear reactors.

Specific problems dealt with in 1994 are:

- The development of collision probability routines for elementary cells of LWRs and their implementation in an interface current flux routine (3.1.1).
- Participation in an international benchmark test, to assess the quality of prolonged burn-up predictions (3.1.2).
- Calculations of the induced radioactivity of some construction elements of the Swedish Ringhals 2 pressurized water reactor (3.1.3).
- Assistance to the operation of the DR 3 reactor (3.1.4).
- Generation of cross sections for heavy nuclides (3.1.5).

3.1.1 Nodal Collision Probability Theory

The neutronics part of an LWR fuel assembly code NICOLAS is under development. The neutron flux distribution for each energy group of the multigroup formalism is based on the flat-flux, isotropic collision probability method within each lattice cell combined with interface-current techniques to represent the cell interactions of the system. The flux solution techniques used in this code are presented in two papers which have been accepted for publication in Nuclear Science and Engineering, March 1995.

The first paper, "Transfer Probability Calculation for Square Pin Cells by Two-Dimensional Gaussian Quadrature" is concerned with the application of very efficient integration techniques for the evaluation of collision, escape, and transmission probabilities for circular pin-type cells with square boundaries. At this stage of development, it was possible to model only regular lattices occurring in pressurized water reactors (PWR).

Recent developments to meet the requirements of boiling water reactors (BWR) encompass subroutines for three more cell types related to water gaps with flow channel wall, corner cells, and control-blade cells. These subroutines cover the most basic BWR features as well as the most regular types of large water holes in modern fuel assembly types (e.g. Siemens' Atrium). Further development will be needed to cope with large circular water holes, power range monitor tubes, and the sophisticated SVEA-96 fuel geometry.

While the cosine current approximation is satisfactory for PWR lattices, it is expected that BWR test calculations using the new subroutines will reveal a need to incorporate first-order Legendre terms to obtain sufficient accuracy even for the more heterogeneous cases.

The solution of the system equations is the subject of the second paper, "Investigation of Interface-Current Solution Techniques for Coupled Heterogeneous Cells". The basic first-flight probability equations are preconditioned by Gaussian elimination of the subregion fluxes, resulting in a set of multiple-flight probability equations. The interface-current equations thus turned into a nodal form are solved iteratively, alternating between successive overrelaxation and free iterations, and using current rebalancing techniques. Finally, the converged currents are used to calculate the fluxes. The paper includes an analysis of the convergence rate in terms of the system matrix properties and describes the results of extensive performance testing to find the optimal iteration strategy.

3.1.2 Participation in an International Benchmark Test

The Nuclear Science Committee of the OECD organizes, among its activities, comparisons of computer code performance.

The Reactor Physics Section has participated in the benchmarks test "Burn Up Credit Criticality", which concerns the prediction of the production/destruction of actinides and fission products during long burn-up periods. The benchmark test is performed to obtain an overview of the theoretical predictions of burn-up calculations in order to allow safe shipments of larger numbers of spent fuel assemblies. At present the spent fuel assemblies are treated as new assemblies, when criticality estimates are made.

A draft report of the results has been issued by the American coordinator. Our results were generally in good agreement with those of the majority of the participants.

3.1.3 Ringhals Calculations

Last year calculations were reported concerning activities of construction elements of the Swedish PWR Ringhals-2. These elements were situated in low-flux regions far away from the reactor core, and streaming in the open space around the pressure vessel was responsible for most of the neutrons. However, streaming effects cannot be properly treated in the diffusion theory approximation, upon which the programs used were based.

New calculations have been performed with a Monte Carlo code, MCNP3, which can treat the problem rigorously. The rather surprising result is that the results of the diffusion calculations are not so bad after all, i.e. in most regions within a factor of 5 from the Monte Carlo results.

3.1.4 DR3 Reactor Physics

The computer code DR3SIM, applied by the operating staff of the reactor DR3, has been modified during 1994 on demand from the user. A more user-friendly input and output interface, especially for withdrawing the stainless steel absorbers used for burn-up compensation, has been implemented. Finally some minor errors have been corrected.

3.1.5 Generation of Cross Sections for Heavy Nuclides

The neutron cross section library presently in use at Risø is based on the UNKDL from the 1970's. It does not contain cross sections for the more exotic heavy nuclides, which are important at very high burn-ups or when transmutation studies are to be performed.

From the data library delivered with the Monte Carlo code MCNP3 (essentially the American evaluated libraries ENDF/B-IV and ENDL-85) a new library in our 76 group structure has been made, which contains data for a large number of the heavy nuclides.

3.2 Reactor Safety

The work has dealt with

- Studies of hydrogen deflagration in containments under accident conditions (3.2.1).
- Collection of reactor data for European reactors (3.2.2)
- Participation in the SIK and RAK projects conducted by the Nordic Nuclear Safety Research Programme (NKS):
  - SIK-2, which addresses "Severe Accidents". The work has been finished, although a final report is still to be issued.
  - SIK-3, where safety-related data for reactors in neighbouring countries are collected. The work has been finished, and a final report issued.
  - RAK, the new reactor safety projects, was started in 1994 (3.2.3).
- Participation in the work of the Nuclear Preparedness Group (3.2.4).

3.2.1 Hydrogen Deflagration

A Ph.D. project considering multipoint ignition of mixtures of hydrogen and air has been completed. It included implementation of a model for simulation of the combustion in the computer program CONTAIN 1.12, experiments performed at the Containment Test Facility at Whiteshell in Canada, and comparison of experiments and calculations. The results are reported in the dissertation "Hydrogen Problems Related to Reactor Accidents", Risø-R-706 (EN).

3.2.2 Collection of Reactor data for European Reactors

On the request of the Danish Emergency Management Agency a data base containing standardized information about all operating European power reactors has been prepared.

Also for the Danish Emergency Management Agency, a generic description of boiling water reactors has been made, suited for presentation on a computer screen.

3.2.3 RAK Projects

In 1994, two nuclear-safety-related four-year projects were started within the NKS programme. The first one (RAK-1) examines whether authorities and the industry cover all essential aspects that need to be taken into account in order to ascertain safety. The second one (RAK-2) deals with tools that are needed when examining the questions of severe accidents.

Within the framework of RAK-1, a Danish proposal of a safety related comparison between an advanced boiling water reactor and a modern Nordic boiling water reactor was postponed, possibly to be accepted later in the four-year project period.

Within RAK-2, a Danish proposal to collect information on reactors in the United Kingdom was accepted. It is a continuation of the previous NKS-project, SIK-3, "Design and Safety Features of Nuclear Reactors Neighbouring the Nordic Countries".

Finally the Reactor Physics Section is also participating in the RAK-2 subproject "Recriticality Studies within Severe Accidents".

3.2.4 Work in the Nuclear Preparedness Group

The nuclear preparedness group has mainly been occupied with the collection of information on the state of nuclear power in the world. A Risø report (in Danish) has been written (to be issued early in 1995) covering the subject in 7 chapters:

- The nuclear electricity production worldwide
- Major safety related incidents in 1994
- Barsebäck and the other Swedish nuclear power plants
- The development in Eastern Europe with respect to reactor safety
- The development in the rest of the world
- The development of reactor types
- The fuel cycle.

Some members of the Nuclear Preparedness Group have conducted a study on the thermal hydraulics of boiling water reactors in order to improve the models used presently for prediction of void distributions. Such an improvement is considered necessary for the treatment of natural circulation BWR's, e.g. the SBWR.

3.3 Activities at the DR1 Reactor

Activities at the DR1 reactor have comprised

- Courses on experimental reactor physics for university students and high-school classes (3.3.1).
- Irradiation of computer memories with fast neutrons to examine bit error frequencies (3.3.2).

3.3.1 Education

The reactor has been used almost exclusively for educational purposes. 49 high school classes have carried out one day or half-a-day experiments at the reactor. The total number of high school pupils in 1994 was about 900.

A number of students from the Technical University of Denmark have carried out experiments at the reactor over a period of three weeks.

Some of the experiments were:

- Determination of the reactor's temperature-, power-, and bubble-coefficients
- Neutron activation analysis
- Measurements of neutron cross sections
- Neutron radiography
- Health physics experiments
- Core flux distribution measurements.

3.3.2 Neutron-Induced Bit Errors in Computer Memories

In 1994, various CMOS-SRAM circuits were tested by neutron irradiation for an electronics company.

Because of small feature size, computer memories are sensitive to neutron irradiation. This can be a problem in computers used on long-distance passenger flights.

The neutrons are generated at flight altitudes when cosmic ray particles hit oxygen or nitrogen nuclei in the atmosphere. When a fast neutron thus generated hits a silicon nucleus, the nucleus will recoil, and it may produce an ionization large enough to cause a bit error in the memory. The error pattern seen at flight altitudes is readily reproduced by exposure of the circuit to fast neutrons from a neutron source available at DR1.

3.4 Publications

3.4.1 Publications in International Journals, Proceedings and Reports

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Nonbøl, E. (ed.), Design and safety features of nuclear reactors neighbouring the Nordic Countries. Final report of the Nordic Nuclear Safety Research Project SIK-3. (The Nordic Council of Ministers, Copenhagen, 1994) (TemaNord, 595) 57 p.

3.4.2 Risø Reports

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Fynbo, P.B., Atomkraftværker - nogle forhold af betydning for deres sikkerhed. Risø-I-827 (1994) vp.

Højerup, C.F., Danish calculations of the NEACRP pin-power benchmark. Risø-R-681 (EN) (1994) 52 p.

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Højerup, C.F., Calculations of neutron fluxes in 'far-away-from the core regions' of the Ringhals-2 reactor by Monte Carlo methods and by diffusion theory. Risø-I-796 (1994) 26 p.

Majborn, B.; Brodersen, K.; Damkjær, A.; Højerup, C.F. (eds.), Nuclear Safety Research Department. Annual progress report 1993. Risø-R-739(EN) (1994) 30 p.

3.5 Lectures at Conferences and Meetings

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4 Waste Management

The Waste Management Plant at Risø takes care of handling and storage of radioactive waste from all Danish users of radioisotopes. Measured as volume, about a third of the waste is coming from outside, primarily from hospitals, industry and various laboratories. However, the activity in the waste is dominated by contributions from the nuclear facilities at Risø.

The storage facilities at Risø are only intended for temporary use and no permissions for permanent disposal of radioactive waste in Denmark has been given by the competent authcrities. As the situation is internationally, it must be expected that Denmark will have to create a permanent repository for at least the low- and medium-level waste. However, for reasons connected with future decommissioning of the major nuclear facilities at Risø, it is suitable to postpone disposal of the stored waste until some time into the next century.

In the meantime members of the staff from the Waste Management Plant are participating in international research concerned with the safety of disposal systems. The work is carried out in connection with Nordic cooperative projects and the EU research programme on radioactive waste management. In the future this work will formally be part of the Radiation Protection Research Programme at Risø, but it will continue to be a major channel through which the staff at the Waste Management Plant is contributing to and keeping contact with the international development within the field of radioactive waste management. Participation in the EU advisory commitee on practical waste management is also valuable in this context.

4.1 Practical Waste Management

The treatment methods for radioactive waste are unchanged from previous years: A balling press is used for compaction of low-level solid waste and a destillation plant based on the steam recompression principle is used for purification of radioactive waste water. Bituminization is employed as conditioning method for the resulting evaporator concentrates.

The Waste Management Plant is also taking care of

- the decontamination and laundry facilities at Risø,
- the collection of chemically toxic waste and transfer of such materials to "Kommunekemi", the central Danish facility for management of such materials, and
- the biological purification system for ordinary sewage water from the Risø area.

Preprojects for general renewal, capacity increase, and reconstruction of the inactive waste water purification system to include nitrogen purification were prepared in 1994 by COWIconsult A/S. The new facility will be built in 1995.

The question about disposal of the very veakly contaminated sewage sludges from the present waste water purification system has still not found a satisfactory solution. Some 50 t (as dry material) is now in interim storage. Permission has been asked from the environmental protection authorities for the use of the controlled waste tip at Risø for disposal of the sludge. Disposal outside of Risø may still be a possibility.

The destilled water from the evaporation plant and a few other minor sources for release of radionuclides are mixed into the main stream of inactive sewage water before purification. The system is monitored for radioactivity at various points and the releases are reported to the nuclear authorities. The total release of mixed β-emitters with the purified waste water was ~ 74 MBq. This is about 0.6 % of the permitted releases to Roskilde Fiord. In addition 11300 GBq of tritiated water were released with the destilled water and about 4 GBq ^{14}C-carbondioxide were released to the atmosphere with off-gas from the bituminization plant. The control and reporting system for the releases were evaluated by an EU inspection team visiting Risø in 1994.

Decommissioning of the Risø Hot Cells was finalized in 1994 and resulted in unexpectedly large volumes of α-contaminated machinery, tools, etc. The waste was delivered packed in steel boxes which had to be stored in the concrete vaults intended for this type of waste at "Centralvejslageret". This and other types of waste units from the Hot Cell decommissioning have occupied nearly all the storage capacity in the facility.

The ordinary radioactive waste treatment procedures resulted in a total of 119 drums containing bituminized evaporator concentrate or compacted low-level solid waste.

The transfer of old waste units from the previous storage area "Betonrørslageret" to the new storage building for low-level waste near the Waste Management Plant was continued in 1994. At the end of the year 1717 drums corresponding to 52 % of the units originally present in Betonrørslageret were removed. About a third of the old units had to be overpacked before positioning in the new storage building. The state of the old waste units varies from very corroded to practically undamaged.

External γ-measurements of radioisotopes are made on all drums placed in the new storage building. Information on the contents of ^{137}Cs and ^{60}Co are obtained. The ^{137}Cs can mainly be ascribed to after-irradiation investigation of spent-fuel pellets at the Hot Cell and can therefore be used to estimate contents of other long-lived radioisotopes in the waste. The assumtion will be checked by plutonium determination in selected samples of old and new waste products. The necessary equipment for α-analyses was purchased in 1994.

4.2 Waste Management Research

In addition to information about the contents of important radionuclides, safe management and disposal of radioactive waste require knowledge about the general properties of the materials present in the waste units. Long-term interaction between various parts of the waste units or between the waste units and surrounding materials is important for safety assessments. Information valid over very long periods is needed and will have to be based on understanding of the fundamental chemistry and physics involved. Staff from the Waste

Management Plant is contributing to waste material research within the following areas:

4.2.1 Properties of Bituminized Materials

Studies under a four year CEC contract on "Characteristics of Bituminized Waste" was continued in cooperation with CEN Cadarache, France, and CEN/SCK Mol, Belgium. The third annual report 1994 is in preparation.

The experiments carried out at Risø are using techniques developed under previous contracts. The following properties are measured:

- Diffusivites of water and ions in thin bitumen membranes as function of content of salt or sludge particles.
- Leaching, water uptake and swelling of bituminized materials containing soluble salts.
- Pressurization of confined samples by water uptake through barriers of porous cement mortar.

A product from the bituminization plant at Risø is included in the present series of experiments. The rates of leaching, water uptake and swelling are found to be rather high, in agreement with other measurements on real waste products. Osmotic pressures higher than 3 bars are reached. Pressures that high cannot be contained by ordinary waste units, and the development of cracks or outflow of bituminized product must therefore be expected. Some phenomena observed in connection with the transfer of old drums with Risø waste can be explained in this manner.

It has been found that the EE SEM instrument (Electroscan Environmental Scanning Electron Microscopy) available at the Materials Research Department is well suited for pictorial documentation of the bubble structure of swelled bituminized material. An example is shown in Figure 4.2.1a. The advantage of EE SEM is that it tolerates wet samples and the slight organic vapour pressure associated with bitumen.

Various models describing the swelling and leaching behaviour of bituminized materials have been developed and descriptions will be published in 1995. The main model simulates primarily the water uptake and swelling of bituminized products containing sodium nitrate crystals. Leaching is also included. The development with time is relatively simple in the idealized case of uniform size crystals placed in a regular grid, but the situation gets much more complex for randomly positioned crystals with a size distribution. This more realistic situation may be handled using another type of model where the simulation of water diffusion is omitted but the likely bubble structure of the swelled product can be presented as function of a selected water content. An example from such a calculation is shown in Figure 4.2.1b. and is seen to be a reasonable approximation to the EE SEM picture in Figure 4.2.1a.

The experiments and modelling was presented at EU working group meetings at Harwell and Aberdeen. The models were demonstrated and discussed with French collegues at Cadarache in the autumn 1994.

4.2.2 Properties of Cementitious Materials

The CEC contract on "The Performance of Cementitious Barriers in Repositories" is a cooperative project between AEA Technology, Harwell, and Risø. Two minor contributions are included in form of tomography supplied by BAM (Bundesanstalt für Materialprüfung), Berlin, and an investigation of the use of radon for characterization of diffusion in water-saturated porous material carried out by NR Rez, the Czech Republic, under the PECO programme. The late inclusion of the last subproject has resulted in postponement of the preparation of the final report which now is expected to be available medio 1995.

The experimental work at Risø stopped in the spring 1994. It was mainly concerned with crack filling and crack surface covering in cementitious barriers due to flow of calcium bicarbonate containing water. The mechanisms involved are now reasonably well understood. Thin cracks or crack systems with many dead-end side cracks show a high tendency to crack closure. However, the

Figure 4.2.1a. EE SEM picture of the bubble structure in a sample of bituminized material after exposure to water in 147 days. Originally the bituminized product contained 10% $NaNO_3$ crystals with sizes between 10 and 100 µm and 30% $BaSO_4$ about 1 µm large insoluble crystals. The cut is perpendicular to the water exposed surface (just visible in the upper right corner). The sodium nitrate crystals have dissolved in the water diffused into the sample, leaving the interconnected solution-filled bubble structure. In the bottom left corner a partly undissolved $NaNO_3$ crystal can still be seen. In the layers below the crystals get more abundant. The barium sulphate represents the insoluble sludge material often present in such waste products. The gray spots are lumps of barium sulphate.

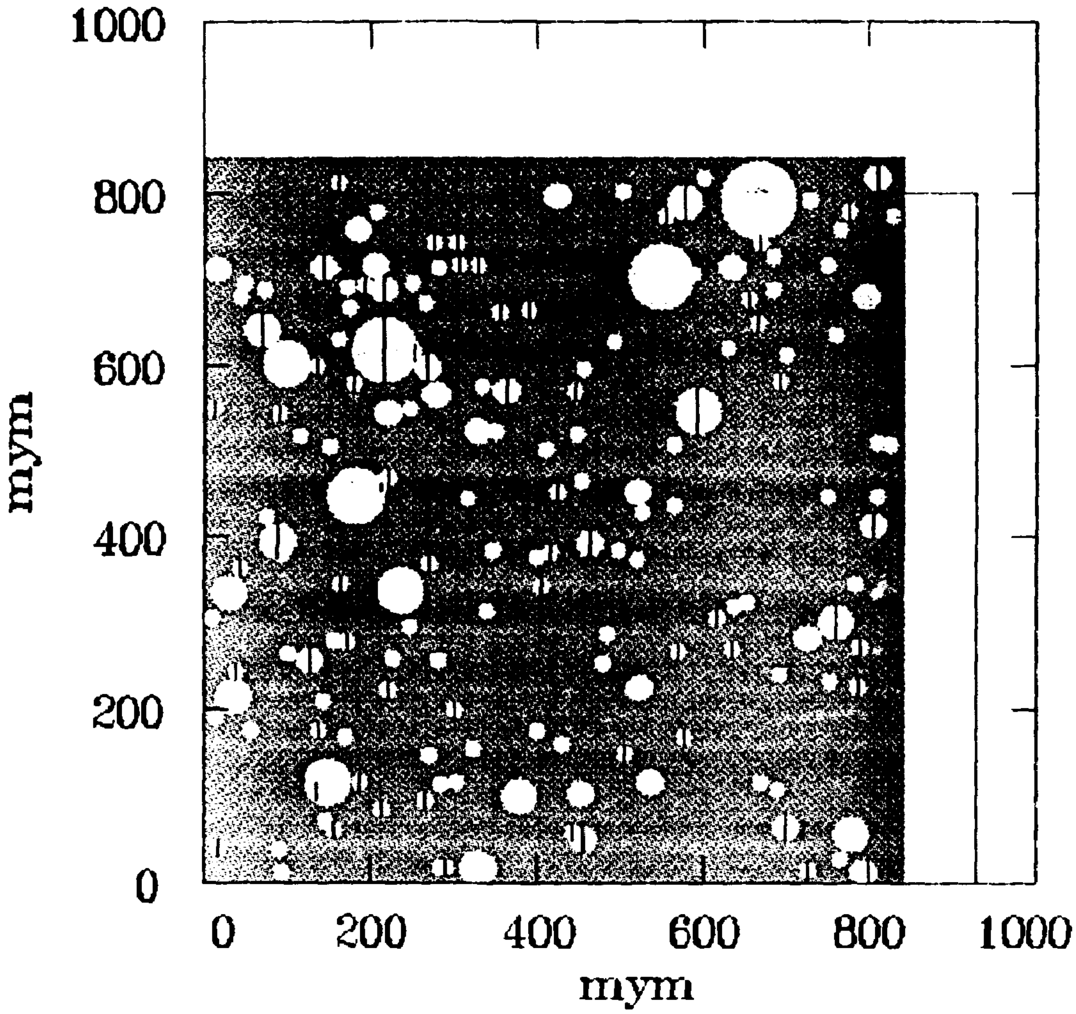

Figure 4.2.1b Model presentation of a cut through a sample of bituminized material. The original sodium nitrate content was 10% in form of crystals with sizes between 10 and 100 μm randomly distributed inside the bitumen matrix. The size of the circles corresponds to bubbles filled with saturated solution containing the same amount of sodium nitrate as the original crystal present at that position. Bubbles with unsaturated solution will be correspondingly larger, as is the case for the bubbles in the upper part of Fig. 4.2.1a.

formation of relatively impermeable layers of deposits on the internal surfaces of coarser cracks are of greater relevance from a safety point of view, because the deposits prevents efficient contact between the flowing solution and the bulk of the cementitious material. Such cracks may then turn into preferential flow channels short-circuiting the barrier system.

The formation of a covering layer of calcium carbonate crystals results in very steep concentration gradients over the layer. A numerical model of the layer formation is available, but the steep gradients and the thin diffusion nodes necessary to describe the layers are still giving problems. Additional work on the model was (and will be) carried out in connection with the writing of the Risø-contributions to the final report for the project.

The measurements made by the Environmental Science and Technology Department using SANS (small angle neutron scattering) to characterize the micro-structure of cement paste have been supplemented by measurements on various pure minerals typical for cement paste cured at different temperatures. The minerals were synthesized at the University of Aberdeen. The SANS results are excellent and may be utilized in a separate paper.

Results from the crack studies were presented at the above mentioned working group meetings at Harwell and Aberdeen. A presentation of results from the study was also included in the NKS meeting on "Radioactive Waste in the Nordic Countries" in Helsinki in April 1994.

The experimental method employed in the crack filling experiments is versatile and various applications will be proposed for the Radioactive Waste part of the new EU research programme on Nuclear Fission Safety.

The migration of chloride ions into concrete and their influence on corrosion of embedded steel is the topic of a working group set up by DBK (Danish Concrete Association). The review report from the group is delayed but participation in the work has given some interesting experiences concerning practical use of concrete.

4.3 Soil Chemistry

Some work on general soil chemistry and ecology is also carried out by the staff at the Waste Management Plant. The main objective is development of a model for the elemental turnover in soils and plants in a forest area. It is made for the NECO project (Nitrogen Deposition, Turnover and Effects in Coniferous Forests Ecosystems) financed by the Danish Strategic Environmental Research Programme.

The SAMUS model (single area model of unsaturated soil) is based on the ECCES model previously developed at Risø. The main effort in 1994 has been the introduction of the nitrogen system into the model, and writing of a general description of the model. According to the contract, considerably more work is expected for 1995.

4.4 Publications

4.4.1 Publications in International Journals, Proceedings and Reports

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Unpublished. Abstract available.

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Appendix 1

STAFF of the Department 1994

Head of Department
Benny Majborn

Office Staff
Inge Blyitgen
Margit Nielsen
Lis Rasmussen

Health Physics Section

Scientific Staff
Anders Damkjær (head of section)
Claus Erik Andersen
Jette Borg (Ph.D.student)
Lars Bøtter-Jensen
Poul Christensen
Ayoe Hoff (Ph.D.student, from March 1)
Niels A. Larsen (Ph.D.student, from March 1)
Jørgen Lippert
Flemming K. Nielsen
Nigel Poolton (post doc. fellow)
Frits Heikel Vinther (until May 26)
Ole Walmod-Larsen

Technical Staff
Birthe Berg
Per Brøns
Henrik E. Christiansen
Lissi Sture Hansen
Jørgen Jakobsen
Nina Jensen
Johannes Jepsen
Finn Jørgensen
Finn Pedersen
Lis Sørensen
Finn Willumsen

Guest Scientists
Steve W.M. McKeever
(August 1 to December 31), Oklahoma State University, USA
Högne Jungner
(March 6-13, May 9-16 and October 6-8), University of Helsinki, Finland
Barbara Mauz
(January 1 to April 30), Max Planck Institute, Heidelberg, Germany

Apprentices
Per Hansen

Reactor Physics Section

Scientific Staff
C.F. Højerup (head of section)
P.E. Becher
A. Bujor (Ph.D.student, until February 28)
Peter Bille Fynbo
Erik Nonbøl
Knud Ladekarl Thomsen

Waste Management Section

Scientific Staff
Knud Brodersen (head of section)
Massimo Steen Carugati

Technical Staff
Birthe Andersen
Winnie Andersen
Jørgen E. Bindner (May 1 - August 13)
Mogens Christiansen
Birthe Hansen
Signe Hansen
Ole Sølling Hansen
Kurt Jensen (temporary)
Fini Jørgensen (temporary)
Gitte Larsen (until February 21)
Jørgen Larsen
Mogens Larsen (temporary, from October 1)
Peter Nielsen
Palle Olsson
Jesper Bohn Rasmussen
Nina Thomsen
Bent Willumsen
Arne Vinther
Ruth Aagesen

Appendix 2

Participation in International Committees.

IAEA, The International Atomic Energy Agency
INES Users Group (Becher)

OECD, Nuclear Energy Agency
CSNI, Steering Committee (Højerup)
CSNI-PWG4, Confinement of Accidental Radioactive Releases (Fynbo)
NEA-NSC, Nuclear Science Committee (Højerup)
NEA-Data Bank Executive Group (Højerup)

EC, European Commission
CGC 6 Nuclear Fuel Cycle (Brodersen)
ACPM for Plan of Action (Brodersen)
Task 3 of the Waste Research Programme (Brodersen)
CGC Nuclear Fission Safety (Majborn)
Reactor Safety Working Group (Becher)
LWR Safety Research Index Group (Nonbøl)
Working Party on Criteria for Recycling Materials from the Dismantling of Nuclear Installations (Carugati)
Group of National Experts on Assistance in the Event of a Nuclear Accident or Radiological Emergency (Nielsen)
Article 37 Group of Experts (Walmod-Larsen)
Expert Group on Transfrontier Emergency Planning (Walmod-Larsen)
Expert Group on Environmental Gamma Monitors (Bøtter-Jensen)
Group of Technical Experts on Radiation Protection Dosimetry (Christensen)
EURADOS, Skin Dosimetry (Christensen)
EURADOS, Environmental Radiation Monitoring (Bøtter-Jensen)

Nordic Co-operation
Steering Committee for NKS Projects (Majborn)
Reference Group on AFA Projects (Brodersen)

Editorial Boards
Radiation Measurements (Bøtter-Jensen)
Radiation Protection Dosimetry (Bøtter-Jensen)
Nuclear Europe Worldscan (Nonbøl)
Standing Committee of the 10th and 11th Solid State Dosimetry Conferences (Bøtter-Jensen)

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Abstract (Max. 2000 characters)

The report describes the work of the Nuclear Safety Research Department during 1994. The activities cover health physics, reactor physics, operation of the small reactor DR1, and radioactive waste management.

Lists of staff and publications are included together with a summary of the staff's participation in international comm[illegible]es.

Descriptors INIS/EDB

INTERNATIONAL COOPERATION; PROGRESS REPORT; RADIATION PROTECTION; RADIOACTIVE WASTE MANAGEMENT; REACTOR PHYSICS; REACTOR OPERATION; REACTOR TECHNOLOGY; RESEARCH PROGRAMS; RISOE NATIONAL LABORATORY

Available on request from Information Service Department,
Risø National Laboratory
(Afdelingen for Informationsservice,
Forskningscenter Risø),
P.O. Box 49, DK-4000 Roskilde, Denmark
Telephone (+45) 46 77 46 77, ext. 4004/4005
Telex 43 116 · Telefax (+45) 46 75 56 27

RISØ

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- *Materials and measuring techniques for industry*

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- *Energy planning*
- *Environmental aspects of energy and industrial production*
- *Environmental aspects of plant production*
- *Nuclear safety and radiation protection*
- *Materials with new physical and chemical properties*
- *Structural materials*
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Key Figures

Risø has a staff of just over 900, of which more than 300 are scientists and 80 are PhD and Post Doc. students. Risø's 1995 budget totals DKK 476m, of which 45% come from research programmes and commercial contracts, while the remainder is covered by government appropriations.

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Available on request from:
Information Service Department
Risø National Laboratory
P.O. Box 49, DK-4000 Roskilde, Denmark
Phone +45 46 77 46 77, ext. 4004/4005
Telex 43116, Fax +45 46 75 56 27